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Lifshitz-Allen-Cahn domain-growth kinetics of Ising models with conserved density

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The domain-growth kinetics of $p=4$-fold degenerate (2×1) ordering in two-dimensional Ising models with conserved density is studied as a function of temperature and range of Kawasaki spin exchange. It is found by computer simulations that the zero-temperature freezing-in behavior for nearest-neighbor exchange is released when next-nearest-neighbor exchange is included. The Lifshitz-Allen-Cahn growth law is obeyed for all temperatures indicating that the density conservation is irrelevant also for $p>2$.

It is now well established[1-17] that the ordering kinetics of Ising models with twofold degeneracy ($p=2$, ferromagnetic or antiferromagnetic) is described by the Lifshitz-Allen-Cahn[18,19] growth law

$$R(t)\sim t^n \tag{1}$$

with $n=\frac{1}{2}$, independent of whether or not quantities other than the order parameter are conserved. $R(t)$ is the average linear size of the ordered domains. In the case of spinodal decomposition and phase separation (ferromagnetic Ising model) with conserved order parameter,[16,12,20-25] the situation is somewhat controversial although there are now strong indications that the $n=\frac{1}{3}$ Lifshitz-Slyozov exponent[26] describes the growth process rather than a logarithmic growth law.[24,25]

The situation is much less clear for nonconserved order parameter in the case of higher-order ground-state degeneracy, $p>d$. First, there is no reliable theory and, second, computer simulations of a variety of two-dimensional Ising models with $p=3$ and $p=4$ give conflicting results regarding the value of n. In this paper we shall be concerned with the ordering kinetics of $(d=2)$-dimensional antiferromagnetic Ising models with nonconserved order parameter and fourfold-degenerate ground-state ordering. In particular, we raise the question whether or not the domain-growth kinetics depends on the conservation of other quantities such as the density ρ. For an antiferromagnet, the density is the bulk magnetization. The two situations of nonconserved and conserved density (corresponding to the dynamical models A and C in the classification by Hohenberg and Halperin[27]) may be modeled in Ising models by Glauber and Kawasaki spin dynamics, respectively.[1] In the case of nonconserved density, computer-simulation studies of growth kinetics of (2×1) ordering $(p=4)$ in Ising models with nearest- and next-nearest-neighbor interactions[28,29] and modulated order $(p=4)$ in axial next-nearest-neighbor Ising (ANNNI) models[30] all lead to the Lifshitz-Allen-Cahn law.[31] In the case of conserved density $(\rho=\frac{1}{2})$, the situation is more unclear. For the $p=4$ ANNNI model,[32] exponent values close to $\frac{1}{2}$ have been reported, but for nearest- and next-nearest-neighbor Ising antiferromagnets[28,33,34] with (2×1) ordering $(p=4)$ and (3×1) ordering $(p=3)$ much lower exponent values (0.25–0.40) are found. These results should, however, be treated with caution since in some cases there are indications of anisotropic growth.[24,32] In other cases, temperature-dependent exponents and crossover to zero-temperature freezing-in were encountered.[28]

The most far-reaching conclusions on the growth kinetics of $p=4$ Ising models with conserved density were drawn by Sadiq and Binder[28] who, on the basis of a very extensive computer-simulation study, found that the late-time growth is characterized by $n\simeq\frac{1}{3}$. These authors argued that such Lifshitz-Slyozov-type kinetics is caused by an excess density in the thermodynamically relevant domain walls and they predicted other models with domain-wall excess density also to have $n\simeq\frac{1}{3}$ (see, however, Ref. 34). According to Sadiq and Binder[28] an excess density in the domain walls will imply that the wall dynamics is controlled by long-range diffusion.

We shall here provide evidence that the domain-growth kinetics of $p=4$ Ising models with conserved density is indeed consistent with the Lifshitz-Allen-Cahn rather than the Lifshitz-Slyozov growth law and demonstrate that the results obtained by Sadiq and Binder[28] are influenced by their special choice of nearest-neighbor Kawasaki spin-exchange dynamics which leads to the peculiar temperature dependence of the effective growth exponent.

We study the two-dimensional square-lattice antiferromagnetic Ising model with isotropic nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions of equal strength (J):

$$\mathcal{H}=J\left[\sum_{i>j}^{\mathrm{NN}}\sigma_i\sigma_j+\sum_{i>j}^{\mathrm{NNN}}\sigma_i\sigma_j\right], \tag{2}$$

where $\sigma_i = \pm 1$ and $J > 0$. The ground state of this model is (2×1) ordering which corresponds to $p=4$. The symmetry is the same as that of atomic oxygen chemisorbed on (110) surfaces of tungsten.[35] The kinetics is governed by Kawasaki spin-exchange subject to the Metropolis Monte Carlo criterion.[36] Exchange between NN as well as NNN sites is allowed. A control parameter $\delta = \nu_{NNN}/(\nu_{NN}+\nu_{NNN})$ monitors the relative NNN exchange frequency ν_{NNN}. The spins are visited randomly. By this process the density is a conserved quantity whereas the order parameter is not. The initial spin configuration is chosen at random corresponding to an initial temperature of $T_i \sim \infty$. Quenches are then performed to temperatures $\tau = T_f/T_c$ below the critical temperature $T_c \simeq 2.10 J/k_B$. The time evolution of the spin configuration is then followed on a time scale given in units of attempted Monte Carlo spin exchanges per site (MCS/S). Different sizes of lattices with $N = L^2$ spins and periodic boundary conditions are considered in order to assess finite-size effects. The main results reported below are derived for $L = 100$ and $L = 200$. Ensemble averages are obtained at each time by averaging over ten independent quenches.

The ordering kinetics is monitored by calculating the dynamical structure function $S(\mathbf{q},t)$ averaged over the two modulated directions, as well as a number of independent measures of time-dependent length scale of the growing domains. These include the inverse excess energy, $\Delta E^{-1}(t) = [E(t) - E(T_f)]^{-1}$, where $E(T_f)$ is the equilibrium energy at T_f, the square root of the intensity, $L(t) = [N^{-1} S(\mathbf{q}_0,t)]^{1/2}$, of the structure factor averaged over the two Bragg points, $\mathbf{q}_0 = (\pi,0)$ and $(0,\pi)$, and the powers, $k_1^{-1}(t)$ and $k_2^{-1/2}(t)$, of the two first moments of the dynamical structure function. Except for $L(t)$, these measures are self-averaging quantities.[28,25]

Our first observation is, in accordance with the findings of Sadiq and Binder,[28] that, when only NN exchanges are allowed ($\delta = 0$), quenches to $T_f = 0$ lead to a frozen-in glassy structure and cease of growth after a short time. However, by extending the exchange to include NNN as well, we discover that the system grows persistently with no tendency to slow down except when slab configurations are generated at late times.[2,6,29] The scaling properties of the growth process are investigated by searching for scaling functions of the type

$$F_m(x) = k_m^{2/m} S(\mathbf{q},t), \quad m = 1,2, \tag{3}$$

where $x = |\mathbf{q}|\, k_m^{-1/m}(t)$ is a time-dependent scaling variable. In Fig. 1 it is shown, for $T_f = 0$ and $\delta = \frac{1}{2}$, that the data for $S(\mathbf{q},t \geq 60)$ are consistent with the existence of such a function $F_2(x)$. A similar statement holds for $F_1(x)$ as well as for other values of δ. The scaling functions are found to be only weakly dependent on temperature. A similar insensitivity to temperature was found by Sadiq and Binder[28] and by other authors for the $p=2$ Ising model.[6,7] Furthermore, we note from Fig. 1 that the high-$|\mathbf{q}|$ tail of the scaling functions, which contains information about the short-distance structure of the domain pattern and is accessible in, e.g., a small-angle scattering experiment, is well described by a Porod-type law $F_2(x) \sim x^{-\omega}$ for $x \geq 1$, with $\omega \simeq 3.0 \pm 0.3$. The

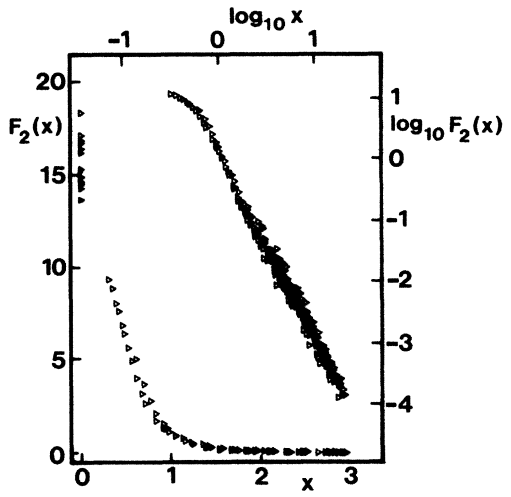

FIG. 1. Zero-temperature dynamical scaling function $F_2(x)$, Eq. (3), in the case of $\delta = \frac{1}{2}$. The data are obtained for a system with 100^2 spins. The time is in units of MCS/S. Only data for $t > 60$ are included in the plot of $F_2(x)$. The data are shown in linear as well as double-logarithmic plots.

value of the decay exponent is consistent with theoretical expectations,[3] $\omega = d+1$, and computer-simulation results for other Ising models.[2,11]

Dynamical scaling, Eq. (3), implies that there is only one relevant length scale of the growth process. This length scale is expected to obey a power law in time, Eq. (1),

$$R(t) \sim L(t) \sim \Delta E^{-1}(t) \sim k_1^{-1}(t) \sim k_2^{-1/2}(t) \sim t^n. \tag{4}$$

Figure 2 demonstrates that this is indeed the case. All

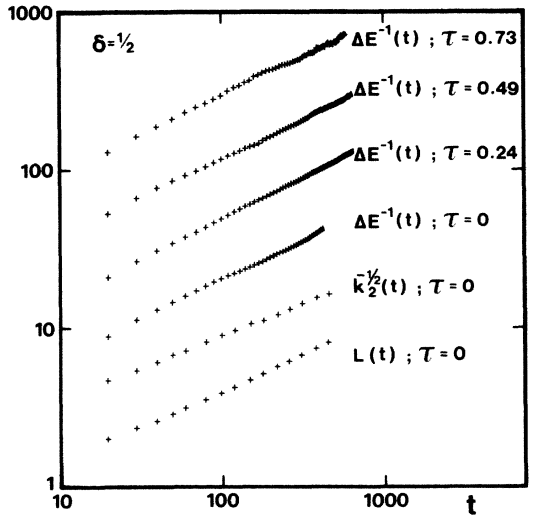

FIG. 2. Double-logarithmic plot vs time of length scales (in arbitrary units) derived from the excess energy $\Delta E(t)$, the second moment $k_2(t)$, of the dynamical structure function, and the amplitude $L(t)$, of the Bragg peak. The data refer to quenches in the case of $\delta = \frac{1}{2}$ for a system with 100^2 spins. The time is in units of MCS/S. $\tau = T_f/T_c$ denotes the reduced quenching temperature.

length measures are described by the same characteristic exponent, the growth exponent n. The crossover to the asymptotic behavior occurs at different times for different quantities. The value of n is close to $\frac{1}{2}$ and we thus conclude that the zero-temperature growth obeys Lifshitz-Allen-Cahn kinetics.

The same relations holds for quenches to finite temperatures, $0 < \tau < 1$, as exemplified in Fig. 2 by $\Delta E^{-1}(t)$. The growth exponent remains at its value close to $\frac{1}{2}$, independent of temperature except in the immediate vicinity of the critical point where the dynamical exponent z has to be introduced in order to analyze the data.[28] Hence, for NN and NNN exchange the Lifshitz-Allen-Cahn growth law describes the computer-simulation data for all temperatures in the ordered phase.

In their computer-simulation study of the model in Eq. (1) with NN exchange only, Sadiq and Binder[28] found that the effective growth exponent exhibits a distinct temperature dependence raising from $n \simeq 0$ at $\tau = 0$ to $n \simeq 0.35$ at $\tau \simeq 0.5$. From $\tau \simeq 0.5$ to 1, n stays approximately constant, $n \simeq 0.35$. In the low-temperature region, the value of n is strongly dependent on the time interval from which the effective exponent value is extracted. These authors then argued that only in the region $\tau > 0.5$ has the asymptotic growth behavior established itself in the available computer simulations. In the light of the results presented in this paper, and considering the lesson learned from other studies of crossover effects due to the influence of freezing-in behavior at low temperatures,[29,37,38] we suggest the following coherent reinterpretation of the computer-simulation results: The exponent values for NN exchange only are influenced by crossover to zero-temperature freezing for all temperatures studied, the influence being stronger the lower the quench temperature. The asymptotic growth region is not reached in the available time span. Longer runs should then show a crossover to the asymptotic behavior characterized by $n = \frac{1}{2}$. However, the asymptotic regime may be reached faster by allowing for NNN exchange. In fact, for equal mixtures of NN and NNN exchange, $\delta = \frac{1}{2}$ (Fig. 2), the asymptotic regime is accessible with modest efforts. These statements are quantified in Fig. 3 which shows results from zero-temperature quenches for varying δ. The inset of this figure gives the resulting late-time exponent as a function of δ.

We then arrive at the simple picture in which the crossover behavior of the Ising model of Eq. (1) is controlled by the parameter δ. For $\delta = 0$, there is freezing-in at

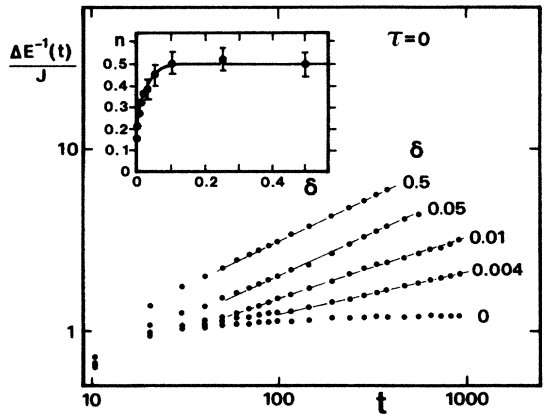

FIG. 3. Inverse excess energy $\Delta E^{-1}(t)$ vs time for zero-temperature quenches for different values of δ. The inset shows the kinetic growth exponent n as a function of δ. The time is in units of MCS/S.

$T_f = 0$. For $\delta > 0$ and $T_f > 0$, the freezing-in behavior becomes unstable and there is a crossover to Lifshitz-Allen-Cahn behavior. The crossover is slower, the lower the temperature and the lower the value of δ. A somewhat similar role was found to be played by the ratio of NN and NNN interactions strengths of the model in Eq. (1) in the case of nonconserved density.[29]

Finally, we wish to comment on some recent experimental results from a low-energy electron-diffraction study of the ordering kinetics of O on W(110).[35] This study is the only one available of the kinetics of (2×1) ordering in a two-dimensional system. In the experiments, the overlayer was prepared in a glassy frozen-in state at low temperatures and then subjected to up-temperature quenches to temperatures $T_f/T_c = \tau \sim 0.3$. The growth was found to be described by a power law with $n = 0.28 \pm 0.05$. For the present chemisorbed system the interaction constants are known[39] and from these we estimate that δ is very small. Hence, the simulation results of this paper suggest that the experimentally found low exponent may suffer from crossover effects due to low-temperature freezing-in behavior.

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